

Gasification of a German Bituminous Coal
With H_2O , H_2 and $\text{H}_2\text{O}-\text{H}_2$ Mixtures

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Bergbau-Forschung has been developing a coal gasification process for a number of years. In this process heat from a high-temperature, gas-cooled nuclear reactor is used as a source of energy for the endothermic steam-carbon reaction (1). All of the coal is thus converted to gas which is a real advantage in countries where the cost of coal is high such as is the case in the Federal Republic of Germany.

As part of the development work on this process, the reaction of several different coal samples with steam, hydrogen and steam-hydrogen mixtures has been investigated extensively in the laboratory. The objective of this work is to develop information needed to design the gasifier used to carry out steam gasification of coal, particularly when hydrogen is present in the system. The results of this investigation with one particular coal, a hard or bituminous coal, is discussed here. During the course of the investigation, samples of this coal were gasified at various steam and hydrogen partial pressures in both pure gases and in steam-hydrogen mixtures. The total system pressure was varied between 1 and 70 atm and runs were conducted over a range of temperatures from 600 to 1100°C.

Apparatus and Procedure

The experimental apparatus has been described previously (2). The coal sample was gasified in a small packed bed reactor that essentially behaved as a differential reactor (Fig. 1). Both isothermal and non-isothermal runs were conducted. During the isothermal runs, the system was heated to the desired temperature using a heating rate of about 200°C/min. The temperature was then held constant. During the non-isothermal runs, the temperature of the system was increased at a constant rate of 10°C/min.

The gasification agent, namely steam, hydrogen or a mixture of both, was fed continuously to the system at a high rate so that the product gas concentration remained small. The products of the reaction were analysed continuously by a mass spectrometer. The apparatus was designed for two different modes of operation, type A for gasification with steam and type B for gasification using steam-gas mixtures.

The coal used was a German bituminous coal, Hagen coal. Analytical data for this coal are shown in Table 1.

Calculational Procedure

The gas analyses from the mass spectrometer were processed directly by a laboratory computer and the carbon conversion was calculated from the amounts of

CH₄, CO and CO₂ in the product gas. Kinetic constants for the gasification rate were determined for each of the gasification agents using the following rate expression:

$$\frac{dX_B}{dt} = k (1 - X_B) \quad 1)$$

where X_B = base carbon conversion as defined by Johnson (3)

t = time

k = kinetic constant

Several other rate expressions were evaluated including the one suggested by Johnson (3). Each of these expressions included an additional term to try to account for the decrease of reaction rate with carbon burn-off. However, none of the expressions fit the data any better than equation 1, so only the kinetic constants for this rate expression are reported here.

Results and Discussion

Steam gasification

Only non-isothermal runs were conducted with steam as the gasification agent, with the temperature of the sample being raised from about 200 to 1100°C using a constant heating rate. Five steam gasification runs were made, 3 at 10 atm and 2 at 40 atm. The main components of the product gas were H₂, CO and CO₂ as previously observed (2). Small amounts of methane also were produced, and the gas composition was not greatly affected by the total pressure.

Since the effects of coal devolatilization on the product gas rate and composition were important up to temperatures between 600 and 700°C, only the data above 700°C were analyzed to obtain kinetic constants. This was done by fitting the integrated form of equation 1 to the base carbon conversion-time data assuming that the Arrhenius equation described the effect of temperature on k . Values of the frequency factor, k_0 , and activation energy, E , were therefore calculated from this procedure. These describe the total rate of base carbon conversion in the reactor. If it is assumed that the carbon dioxide is produced via the gas-phase water gas shift reaction, then these parameters describe the rate of the carbon-steam reaction. However, there was no evidence to indicate that this was the true reaction sequence.

The fit of equation 1 to the carbon conversion data was excellent, as was true of any of the equations used, and a correlation coefficient greater than 0.99 was obtained for each run. Average values of k_0 and E for the two pressures used are shown in Table 2.

Table 2. Kinetic parameters for steam gasification of Hagen coal.

Pressure, atm	k_0 , 1/min	E , kcal/mol
10	3.8×10^4	32.0
40	1.3×10^3	23.0

Examination of the kinetic parameters shown in Table 2 indicates that steam pressure affects the gasification rate. The kinetic constant for steam gasification, k_{H_2O} , was related to both steam pressure and temperature by the following equation:

$$k_{H_2O} = \frac{1.88 \times 10^6 \exp(-2.24 \times 10^4/T) P_{H_2O}}{(1 + 1.56 \times 10^5 \exp(-1.65 \times 10^4/T) P_{H_2O})} \quad 2)$$

Several runs were made with Hagen coal at pressures less than 10 atm. Results from these runs were not adequately described by equation 2, with the indication being that the steam pressure had only a small effect on gasification rate at pressures less than 10 atm.

Hydrogasification

There seems to be general agreement that hydrogasification of coal occurs in three stages, devolatilization, rapid-rate methane formation and low-rate gasification. If the coal sample is heated rapidly to temperatures above 800°C, devolatilization and rapid-rate methane formation occur rapidly and together, and are followed by low-rate gasification (3). Below 800°C, rapid-rate methane formation is slower and occurs over an extended period of time and is therefore more difficult to separate from the low-rate gasification period. Results from the gasification of Hagen coal with hydrogen qualitatively fitted this description of hydrogasification. To determine the rate of hydrogasification of Hagen coal, a series of isothermal experiments were conducted at 10 and 70 atm and at temperatures between 600 and 1150°C. Because of the initial heating period, it was difficult to draw conclusions about the period of rapid-rate methane formation. However this period lasted for less than 15 minutes once the sample reached the reaction temperature and therefore data taken after this period were used along with equation 1 to determine kinetic constants for low-rate hydrogasification. The product gas was generally pure methane during this period, indicating that the only reaction taking place in the system was that between C and H₂ to form CH₄. In a few of the runs, particularly at 10 atm and temperatures of 700°C or below, the product gas contained some CO.

The correlation coefficients obtained with equation 1 were again greater than 0.99 for all runs. The gasification rate seemed to decrease slightly with increasing carbon conversion, but equation 1 still represented the data as well as any of the equations tried. Values of k_0 and E for the two pressures used were derived from the values of the kinetic constants and are shown in Table 3.

Table 3. Kinetic parameters for hydrogasification of Hagen coal.

Pressure, atm	No. of runs	k_0 , 1/min	E, kcal/mol	Correlation coeff.
10	10	1.28	15.6	-0.987
70	14	32.1	17.8	-0.978

The hydrogen pressure strongly affects the hydrogasification rate. The effect of temperature and pressure were correlated using equation 3, which is similar in form to equations suggested by several other investigators (3,4).

$$k_{H_2} = \frac{0.00402 \exp(-5200/T) P_{H_2}^2}{1 + 0.00648 \exp(4100/T) P_{H_2}} \quad 3)$$

Gasification with Steam-H₂ Mixtures

To determine the effect of hydrogen partial pressure on the rate constant for steam gasification, a series of isothermal runs was carried out over the temperature range from 700 to 1100°C. Total pressures of 10 and 40 atm were used, with hydrogen partial pressures between 1.7 and 32 atm.

Equation 1 was again used to determine kinetic constants from the base carbon conversion-time data. Data taken during the run after the first 15 minutes at temperature were used in the calculations so as to avoid the effect of the rapid-rate methanation period. Correlation coefficients greater than 0.99 were obtained for all runs.

Several investigators have treated the kinetic data from coal gasification with H₂-H₂O mixtures by assuming that three basic reactions were occurring (3,5,6). However, to simplify the calculational procedure, it was assumed that the only reactions occurring with Hagen coal were the two that would occur in either pure steam or hydrogen. The rate of steam gasification in the H₂-H₂O mixture, then, assuming that the rate of hydrogasification is independent of steam partial pressure, was the difference between the measured rate constant and the rate constant for the carbon-hydrogen reaction under the same operating conditions. The latter was calculated from equation 3 and was generally less than 10% of the measured rate constant.

The data taken at various hydrogen and steam partial pressures and at 800 and 900°C were examined and the following equation was derived to describe the effects of temperature and the partial pressure of each gas.

$$k_{H_2-H_2O} = \frac{1.88 \times 10^6 \exp(-2.24 \times 10^4/T) P_{H_2O}}{(1 + 1.56 \times 10^5 \exp(-1.65 \times 10^4/T) P_{H_2O} + 3620 \exp(-9830/T) P_{H_2})} \quad 4)$$

Temperature has the largest effect on the gasification rate in steam-hydrogen mixtures. In fact, if the effect of steam and hydrogen partial pressure are ignored, the kinetic constants can be fitted with the Arrhenius equation with a correlation coefficient of -0.904. The resulting activation energy is 33,400 cal/mol and the frequency factor is 2.51×10^4 1/min.

Conclusions

The rate of gasification of a bituminous coal sample in steam, hydrogen and steam-hydrogen mixtures has been measured and the rates correlated by assuming that the base carbon conversion rate was proportional to the amount of base carbon present. Expressions were then found to describe the effects of temperature and steam and hydrogen partial pressures on the rate constants for the steam-carbon and hydrogen-carbon reactions.

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Table 1. Coal analyses.

	Hagen
<u>Proximate analysis, wt.%</u>	
Volatile Matter (maf)	39.2
Moisture	3.7
Ash (mf)	6.3
<u>Ultimate analysis, wt.%</u>	
Carbon (maf)	82.0
Hydrogen (maf)	5.0
Oxygen (maf)	8.2
Nitrogen (maf)	1.5
Sulfur (mf)	2.7

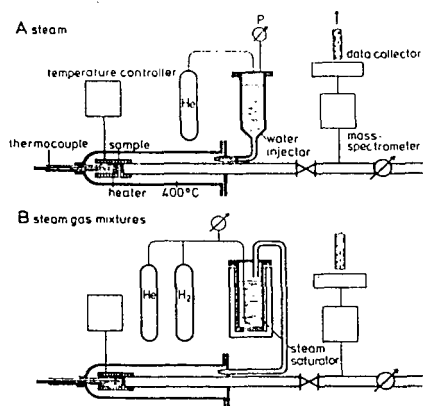


Figure 1. Experimental Apparatus.

Steam gasification of coal in an internally heated fluidized bed

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Abstract

A process is being developed for the steam gasification of coal using heat from a high temperature, gas cooled nuclear reactor. To produce the information needed to design such a system, a 10 cm diameter, internally heated fluidized bed was constructed and has been in operation for several years. A variety of coals have been gasified with steam in this device at pressures up to 40 bar and temperatures between 700 and 900 °C.

Gas compositions, gas yields, heats of reaction and steam decompositions have been determined in the fluidized bed as a function of the operating conditions. Carbon gasification rates have been derived from this data and the variation in gas composition has been correlated using "temperature approach" functions. The effect of the operating conditions on fluidized bed density and heat transfer coefficient also has been investigated.

1. General considerations to an allothermal process

Bergbau-Forschung GmbH Essen develops a process for steam gasification of coal by using process heat from high temperature nuclear reactors. This requires research and development in the field of allothermal gas generators. The envisaged allothermal gas generator is heated by an internally mounted bundle of heat exchanging tubes which are flown through by the gaseous reactor coolant helium. As the helium pressure amounts to 40 bar the steam

gasification is performed at the same pressure. To attain good heat transfer the heat exchanger is located within a fluidized bed of coal and steam {1,2,3}. The coal throughput of such an allothermal gas generator is determined by the heat balance (fig. 1). For a first evaluation of the heat balance assumptions had to be made concerning

- the reaction heat and its dependence on gas analysis,
- the kinetic constants of different feedstocks {4,5,6},
- the heat transfer from the heat exchanger into the fluidized bed, and
- the density of the fluidized bed at high temperatures and pressures.

In the meanwhile experiments have been carried out to measure the above mentioned quantities and their dependence on operating conditions as temperature, pressure, and steam excess.

2. Experimental

Bergbau-Forschung GmbH operates since 1973 a small scale pilot plant in which coal or carbonized coal is gasified in an internally heated fluidized bed at high temperatures and pressures (approx. 1 kg/h). The experimental set up is given in fig. 2. Steam is raised, superheated electrically and fed through a grid into the reaction volume, where a fluidized bed is maintained. . The feedstock is dosed from above by means of a rotating pocket type valve. The crude gas is then passed through a cyclon and a ceramic filter for the separation of particles is then cooled with air, depressurized, dried, volumetrically metered and continuously analyzed by mass spectrometer.

The reaction volume of the gas generator is cylindrically shaped (\varnothing 100 mm, heated height 400 mm). The reaction heat necessary for endothermal steam gasification is generated by an electrically heated metallic spiral. This so called immersion heater is mounted within the fluidized bed in direct neighborhood to the insulation. The diameter of the spiral (80 mm) corresponds to the distance of the heat exchanging tubes in a large scale gasifier.

Fig. 3 illustrates how the heat transfer from the heat exchanger into the fluidized bed is measured. A top view of the reaction volume and the heat exchanger is given, the latter one has a total surface area A and generates an electrical power P . By two thermocouples, one of which has direct contact with the heat exchanger and the other one is mounted on half of the radius, an effective temperature difference can be determined.

The experiments already performed have shown, that just the temperature at half the radius can be used as first order approximation for the mean temperature of the fluidized bed. The heat transfer coefficient α is then calculated by the equation given in fig. 3: $P = \alpha \cdot A \cdot (T_1 - T_2)$.

The devices for determination of height and density of the fluidized bed are shown schematically in fig. 4. By means of three pressure measuring tubes, the first of which is mounted directly above the grid, the second 180 mm above and the third in the freeboard two pressure differences are given. As the measured pressure difference Δp_1 belongs to the known height h_1 the actual height h of the fluidized bed can be calculated. Using then the formula for the hydrostatic pressure the density of the fluidized bed follows.

3. Results

3.1. Gas composition and thermodynamic calculations

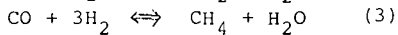
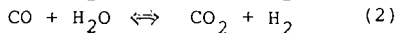
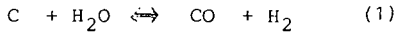
During a run an on-line process computer calculates for instance [7]:

- the frequency of coal feeding,
- the content of the gas generator,
- the absolute carbon conversion,
- the relative carbon conversion,
- the composition of the dry product gas,
- the steam velocity in the gasifier,
- the molar steam carbon ration in the gaseous phase
- the steam decomposition.

Fig. 5 shows data concerning gas production and gas composition during the first 500 minutes of a run at 40 bar and 750 °C with carbonized high volatile bituminous coal "Leopold". The particle sizes used were $< 0,5$ mm. The lower part of the picture shows that after about 200 minutes the gas composition has been essentially constant and the dry product gas consisted of about 50 % H_2 , 26 % CO , 16 % CO_2 and 8 % CH_4 .

Runs with carbonized Rhenish lignite show more CO_2 and H_2 but less CO and CH_4 . The reason for this almost complete conversion of CO to CO_2 which is to be expected for thermodynamic reasons is the relatively high amount of catalytically effective substances in the mineral matter of Rhenish lignite which increases the reaction rate on the one hand and shifts the gas composition more toward thermodynamic equilibrium on the other hand.

The thermodynamics of steam gasification of coal in the absence of free oxygen can be described by three linearly independent reactions, as for instance:



The homogeneous system is determined by two linearly independent reactions as for instance (2) and (3).

Fictitious equilibrium constants can then be calculated from the crude gas as for instance for reaction (3)

$$K_p(\hat{T}_G) = \frac{P_{CO} \cdot P_{H_2}^3}{P_{CH_4} \cdot P_{H_2O}} \quad (4)$$

and lead to fictitious equilibrium temperatures (8). Doing pure thermodynamic calculations the equilibrium temperatures for all reactions used and the reaction temperature coincide. To account for deviations from equilibrium one has to allow different equilibrium temperatures \hat{T}_G . These temperatures \hat{T}_G are generally not correlated with each other and do not coincide with the reaction temperature \hat{T}_R . The difference

$$\Delta \hat{T} = \hat{T}_G - \hat{T}_R \quad (5)$$

is called approach-value of the individual reaction and does depend implicitly on pressure, steam-carbon-ratio, feedstock and experimental set-up.

The gas composition in a system at thermodynamic equilibrium is given by reaction temperature and pressure. At non-equilibrium the approach-values for each of the linearly independent reactions at operating conditions must be known additionally. This

knowledge is now available for some coals being gasified in our small scale pilot plant and is used not only for the representation of the experimental results, but also for the prediction of operating conditions in large scale gas generators.

Some results of our calculations are given in fig. 6. Fig. 6a shows the CH_4 -content in the dry product gas as a function of the steam-carbon-ratio and the reaction temperature. It follows that methane contents of 15 % will be achieved at steam-carbon-ratios of 10 and reaction temperatures of 800 °C, whereas the methane content decreases to values smaller than 1 % with higher steam-carbon-ratios and higher temperatures. As methane formation is strongly exothermal the overall steam gasification reaction heat decreases with increasing methane content in the product gas. According to fig. 6b the reaction heat at low temperatures and low steam-carbon-ratios is less than 1000 kcal/kg feedstock, whereas at high steam-carbon-ratios and high temperatures it attains about 1500 kcal/kg feedstock. In fig. 6c data for steam decomposition are given. It follows that a steam decomposition of about 10 % and more can be attained in our small scale pilot plant at steam-carbon-ratios of 10 and gasification temperatures of 800 °C. A conservative extrapolation of these data from the height of the fluidized bed used here (40 cm) to the height of a fluidized bed in a commercial scale gasifier (3 m) shows that degrees of steam decomposition of more than 30 % can be expected.

3.2 Reaction kinetics and mean gasification temperature

The reaction rate constant used here indicates what percentage of carbon in the solid phase is converted into the gaseous phase by heterogeneous reactions as for instance by (1). The reaction rate defined this way and all kinetic constants derived from it depend implicitly on the composition of the gaseous phase due to the coupling between heterogeneous and homogeneous reactions.

Fig. 7 shows data concerning the reaction rate constants at various temperatures using carbonized high volatile coal "Hagen" as feedstock. At temperatures between 750 and 860 °C the reaction rate constants in the fluidized bed vary between 0,5 and 4,2 % C/min. In a laboratory scale fixed bed differential reactor slightly higher values were measured. The following explanation is suggested: In a differential reactor the concentration of product gas components is negligible and therefore no backward reactions take place. In the upper layers of the fluidized bed, however, hydrogen in the range of some percentages is present. Therefore the reaction rate constants from the upper part of the fluidized bed should be compared with corresponding laboratory values using mixtures of steam and hydrogen as gasifying agent. Experiments carried out under this aspect showed that using carbonized high volatile coal the reaction rate decreases when hydrogen is added (9).

In a laboratory scale differential reactor the gasification temperature can be determined by a single thermocouple. In contrast to this a three-dimensional temperature field exists in the interior of the fluidized bed reactor used here. 12 thermocouples are used to determine this field. Fig. 8 shows the temperature variation 185 mm above the grid. The temperature is the highest in the neighborhood of the electrically heated spiral and decreases towards the center of the fluidized bed. The decrease is especially marked at low steam velocity. Using particle sizes between 0,1 and 0,5 mm a steam velocity of 11 cm/sec is just sufficient for fluidization whereas the fluidized bed collapses at steam velocities of 8 cm/sec. Experiments with even smaller particle sizes showed that using a sieve fraction < 0,1 mm steam velocities of 3 cm/sec are sufficient for fluidization at 40 bar.

Concerning the calculation of a mean temperature of the fluidized bed a first approximation is the integration of the temperature distribution over the reaction volume. Thus a geometric mean value is received which does not take into account that the gasification temperature depends exponentially on temperature and thus the volume elements in the neighborhood of the heat exchanger add more to the gasification throughput than the colder elements in the center of the fluidized bed. Therefore a logarithmic averaging has to be performed for which the activation energy must be known. The activation energy, however, can only be determined when the logarithmic gasification temperature is known. This problem is solved by iteration.

3.3 Heat transfer

The steam velocity does influence not only the horizontal temperature profiles, but also the heat transfer from the heat exchanger into the fluidized bed. In fig. 9 steam velocity w and heat transfer coefficient α are given during an experiment at 40 bar and 830 °C. During this run carbonized high volatile bituminous coal "Leopold" smaller 0,1 mm was gasified. The heat transfer coefficient α was calculated according to fig. 3. Between 70 and 270 minutes and using a steam velocity of about 4 cm/sec the heat transfer coefficient decreased from 1200 to 900 kcal/m²h°C. This can be attributed to an enrichment of ash within the gas generator. The division of α into a radiative and a convective part shall not be discussed here. In fig. 10 the dependance of the heat transfer coefficient α on steam velocity w is shown for two different particle sizes. The steep increase and the subsequent maximum is known {10, 11}. Our experiments at high temperatures and high pressures, however, do not cover the region at very high steam velocities and the transition between fixed bed and fluidized bed at low steam velocities. Our

experiments show as a peculiarity that α_{MAX} is smaller for smaller particles. This can be explained by heat exchanger fouling.

3.4 Density of the fluidized bed

Fig. 11 shows the time dependance of the pressure differences Δp and Δp_1 during a run at 40 bar using carbonized high volatile bituminous coal. Both pressure differences increase simultaneously in the beginning till height h_1 is reached, then Δp_1 remains essentially constant, whereas Δp increases further due to feeding. As soon as the desired height of the fluidized bed is attained the feeding is stopped. Then Δp decreases slowly due to gasification and carry-over. The pressure difference Δp is thus direct information concerning the height of the fluidized bed and can be used for controlling the frequency of feeding. The evaluation of the pressure differences according to the equations given in fig. 4 show that the density of the fluidized bed using carbonized high volatile bituminous is in the range of 300 kg/m^3 .

4. Conclusion

Experiments performed in a small scale pilot plant are used for the determination of

- heats of reaction,
- kinetic constants,
- heat transfer coefficients,
- densities of the fluidized bed.

Though further experiments have to be performed the data already existing are used as basis for extensive computer studies and sensitivity analyses, which are necessary for the design and optimization of a future large scale gasification plant in combination with a high temperature nuclear reactor. All results received till now prove the feasibility of steam gasification of hard coal using nuclear heat at a temperature level of 950 °C.

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Heat Consumed = Heat Transferred

$$q \cdot k_0 \cdot e^{-E/RT} \cdot \gamma \cdot V = h \cdot F \cdot \Delta(T, T_1, T_2)$$

q =	heat of reaction	Gcal/t
k ₀ =	frequency factor	1/h
E =	activation energy	kcal/mol
R =	gas constant	kcal/mol °C
T =	gasification temperature	°K
γ =	density of the fluidized bed	t/m ³
V =	volume of the fluidized bed	m ³
h =	overall heat transfer coeff.	kcal/m ² h °C
F =	heat transferring area	m ²
Δ	log. temperature difference	°K
T ₁ =	helium inlet temperature	°K
T ₂ =	helium outlet temperature	°K

Fig. 1: Heat balance of an allothermal gas generator

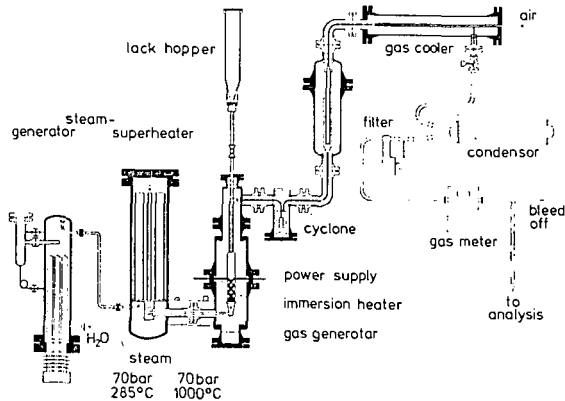


Fig. 2: Small scale pilot plant for steam gasification of coal

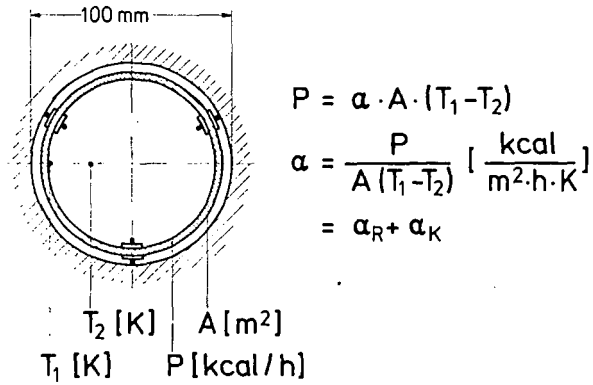


Fig. 3: Determination of the heat transfer coefficient from the heat exchanger into the fluidized bed

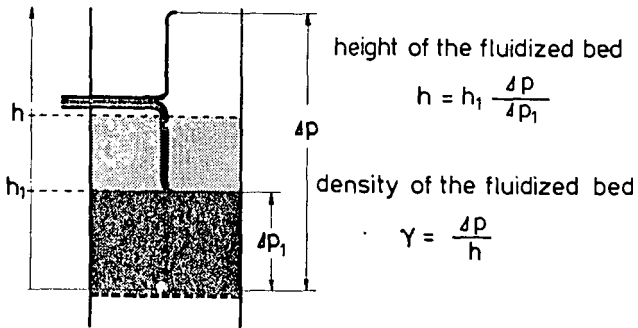


Fig. 4: Determination of the density of the fluidized bed

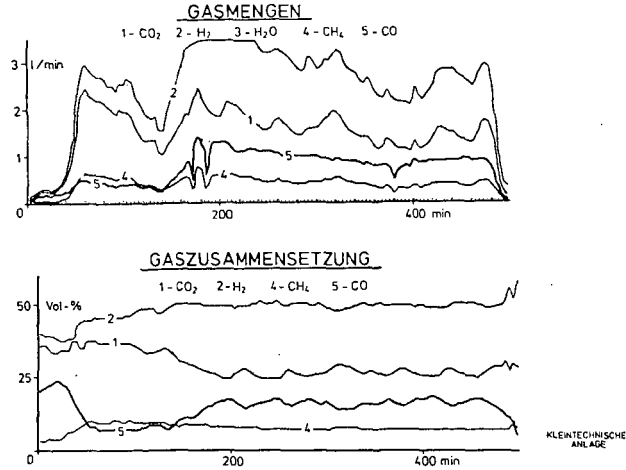


Fig. 5: Steam gasification of carbonized high volatile bituminous coal - gas production and gas composition -

$\frac{H_2O}{C}$ ϑ_R	10	30	100	150	CH₄ Vol % wf
700	27,57	23,27	12,40	8,84	
750	21,83	17,15	7,21	4,41	Schwelkokoks Leopold 40 bar
800	16,36	11,67	3,55	1,78	
850	11,47	7,18	1,45	0,61	

Fig. 6: CH₄-content in the dry product gas (a), reaction heat (b), and steam decomposition (c) as function of reaction temperature and steam carbon ratio

$\frac{H_2O}{C}$ S_R	10	30	100	150
700	338,7	374,6	733,1	916,3
750	528,9	591,7	1021,0	1209,8
800	748,8	842,8	1283,4	1427,6
850	988,5	1104,9	1465,0	1541,1

Utility

kcal/kg Koks

Schwelkoks
Leopold
40 bar

Fig. 6b

$\frac{H_2O}{C}$ S_R	10	30	100	150
700	10,646	3,915	1,387	0,979
750	11,044	4,169	1,506	1,058
800	11,514	4,452	1,610	1,114
850	12,037	4,738	1,679	1,141

WDZ

%

Schwelkoks
Leopold
40 bar

Fig. 6c

Nr	Dauer h	T _{WB} °C	Umsatz %/min
2	5	750	0,5 (0,5)
3	23	800	1,9 (1,7)
4	9	860	4,2 (6,0)
7	18	820	2,5 (2,7)

Fig. 7: Comparison of reaction rate constants measured in a small scale pilot plant and in a fixed bed differential reactor ()

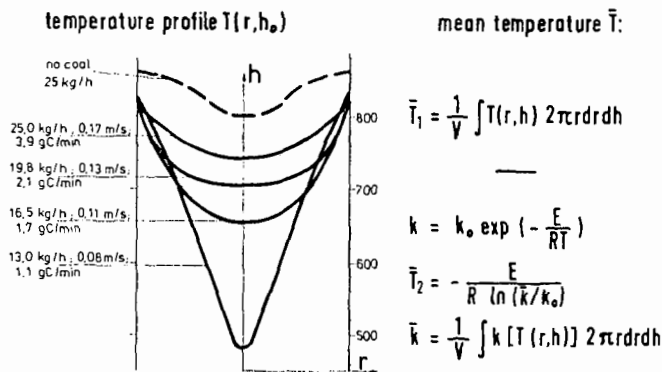


Fig. 8: Temperature profiles in the fluidized bed of the small scale pilot plant

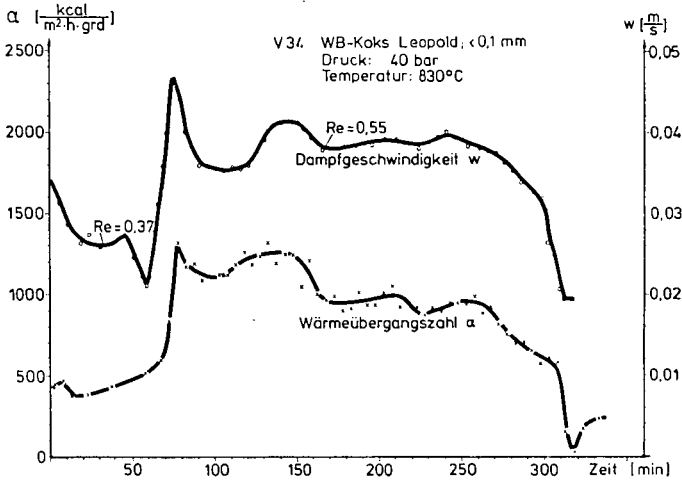


Fig. 9: Heat transfer coefficient and steam velocity as a function of time during a run at 40 bar and 830 °C

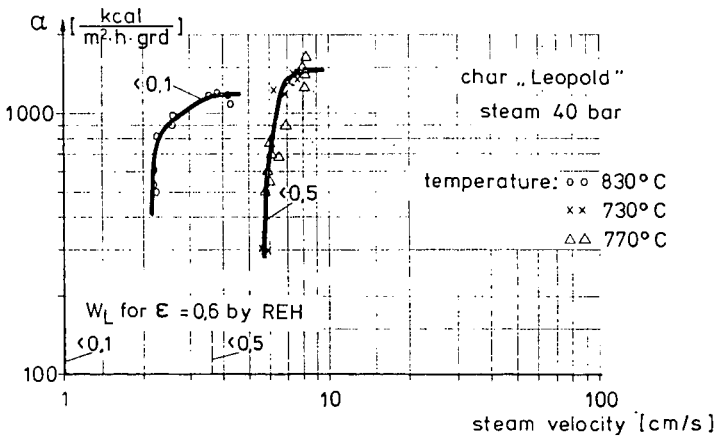


Fig. 10: Heat transfer coefficient as a function of steam velocity using two different particle sizes

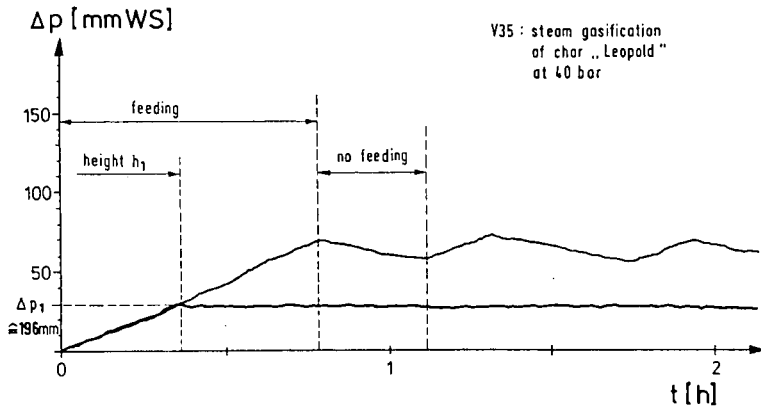


Fig. 11: Pressure differences within the fluidized bed as a function of time